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### **III. REMARKS**

#### **RESPONSE TO REJECTIONS**

##### **In the Claims:**

- **Claim Rejections under 35 U.S.C. § 103(a):**

- **The Examiner's Position:**

Claims 1-3, 13-17, and 18 are rejected under 35 U.S.C. 103(a) as being obvious over US 4,409,414 to Langer, Jr. ("Langer"). Specifically, the Examiner alleges that Langer discloses a batch/continuous process of making alpha-linear olefins of the claimed range of number of carbons by oligomerizing ethylene in the presence of a catalyst containing zirconium alkoxide such as  $Zr(OBu)_4$  and alkyl aluminum halide, in the presence of a diluent such as toluene under the condition of a temperature ranging from below 125°C, a pressure ranging from above 50 psia such as 500 psia, during the claimed time and high-speed stirring (the abstract; col. 2, line 18 thru col. 6, line 34; col. 7, lines 12-29; examples, and the entire reference for details).

Accordingly Langer's process is operated in the presence of alcohol to enhance the polymerization process (col. 5, lines 55-68). On column 6, lines 1-4, Langer disclose ratio of the amount alcohol and the alkyl group of aluminum alkyl. Since the Examiner cannot compare this ratio with the applicant's claimed ratio as called for in claim 1, the Examiner assumes that the ratio used by Applicant is different from the Langer's one. However, as disclosed by Langer, the amount of alcohol effect to the molecular weight of the product (col. 5, lines 55-57).

It would have been obvious to one having ordinary skill in the art at the time the invention was made to have modified the Langer process by selecting an appropriate amount of

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the added alcohol such as the applicants' claimed one according to the desired molecular weight of the product since it has been held by the patent law that the selection of reaction parameters such as temperature and concentration would have been obvious. More particularly, where the general conditions of the claimed are disclosed in the prior art, it is not inventive to discover the optimum or workable ranges by routine experimentation. *In re Aller* 105 USPQ 233, 255 (CCPA 1955). *In re Waite* 77 USPQ 586 (CCPA 1948). *In re Scherl* 70 USPQ 204 (CCPA 1946). *In re Irmischer* 66 USPQ 314 (CCPA 1945). *In re Norman* 66 USPQ 308 (CCPA 1945). *In re Swenson* 56 USPQ 372 (CCPA 1942). *In re So/a* 25 USPQ 433 (CCPA 1935). *In re Dreyfus* 24 USPQ 52 (CCPA 1934).

Langer does not disclose the speed of agitator in the stirred tank. However, Langer discloses operating the reaction by a high-speed stirring (example 1). Therefore, it would have been obvious to one having ordinary skill in the art at the time the invention was made to select an appropriate stirring speed such as 300-1000 rpm to well-mix the reaction as taught by Langer to arrive at the applicants' claimed process.

Claim 19 is rejected under 35 U.S.C. 103(a) as being unpatentable over Langer in view of US 5,260,500 to Shiraki et al. ("Shiraki"). Langer discloses a process as discussed above.

Langer does not disclose adding thiophene into the catalyst (see the whole patent to Langer for details). However, Shiraki discloses that in a process for producing a linear alpha olefins, it is effective to add to the catalyst a sulfur compound such as thiophene to improve the purity of the linear alpha olefins (col. 1, lines 19-28).

It would have been obvious to one having ordinary skill in the art at the time the invention was made to have modified the Langer process by adding an amount of thiophene to

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increase the purity of the product.

Claims 4-12, 20-22, 24-27, and 29 are rejected under 35 U.S.C. 103(a) as being obvious over the cited primary reference to Langer in view of US 4,855,525 to Young et al. ("Young")

While Langer allegedly discloses a process as discussed above, the Examiner admits that Langer is silent on aluminum compounds as called for in claims 4-12, 20, 21, 25, and 26. However, the Examiner contends that Young compensates for the deficiency in Langer by disclosing aluminum compounds such as  $R_3Al_2X_3$ ,  $AlR_2X$ ,  $AlR_3$ , and  $AlRX_2$  as equivalent components for oligomerizing catalysts with X being Cl, R being ethyl (the abstract; col. 4, lines 46-51). Therefore, the Examiner alleges that it would have been obvious to one having ordinary skill in the art at the time the invention was made to have modified the Langer process by using  $R_3Al_2X_3$  and  $AlR_3$  as the aluminum component for the Langer catalyst since it is expected that using any equivalent aluminum compounds disclosed by Young would yield similar results.

Claims 23 and 28 are rejected under 35 U.S.C. 103(a) as being obvious over the combined Langer and Young disclosures further in view of Shiraki.

Langer and Young disclose a process as discussed above wherein the Examiner finds that neither Langer nor Young disclose adding thiophene into the catalyst (see the whole patent to Langer for details), however, alleges that Shiraki compensates for this deficiency by disclosing a process for producing a linear alpha olefins, wherein a sulfur compound, such as thiophene, added to the catalyst to improve the purity of the linear alpha olefins (col. 1, lines 19-28).

In the Examiner's opinion it would have been obvious to one having ordinary skill in the art at the time the invention was made to have modified the Langer process having been

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modified by Young's aluminum compounds by adding an amount of thiophene to increase the purity of the product.

Contrary to Applicant's argument that Langer discloses an *in situ* reaction of the transition metal halide with an alcohol as not persuasive since in the Examiner's opinion Langer allegedly discloses a catalyst including all of components recited in the present claims. The scope of Applicant's claims allegedly does not cover how the catalyst is prepared. Instead, Applicant's claims only what is contained in the catalyst.

According to the Examiner, the argument that *in situ* generated  $\text{ZrCl}_4$  and  $n\text{-BuOH}$  is different from the defined  $\text{Zr}(\text{Obu})_4\text{XbuOH}$  (where  $X=0.3, 0.4, 1$ ) is not persuasive since the catalyst of Langer is formed also by  $\text{Zr}(\text{Obu})_4$  and alcohol. Furthermore, the Examiner contends that Applicant does not claim a catalyst having such formula.

The admitted deficiency of  $\text{AlEt}_2\text{Cl}$  and/or  $\text{AlEtCl}_2$  in Langer is allegedly remedied by Young.

The Examiner also contends that there is motivation to combine Young with Langer, since the Young reference allegedly discloses aluminum compounds such as  $\text{R}_3\text{Al}_2\text{X}_3$ ,  $\text{AlR}_2\text{X}$ ,  $\text{AlR}_3$ , and  $\text{AlRX}_2$  as equivalent components for oligomerization catalysts with X being Cl, R being ethyl (the abstract; col. 4, lines 46-51). In the opinion of the Examiner therefore, it would have been obvious to one having ordinary skill in the art at the time the invention was made to have modified the Langer process by using  $\text{R}_3\text{Al}_2\text{X}_3$  and  $\text{AlR}_3$  as the aluminum component for the Langer catalyst in the expectation that using any equivalent aluminum compounds disclosed by Young would yield similar results. In addition, the Examiner contends that Shiraki uses the sulfur compound to purify the product.

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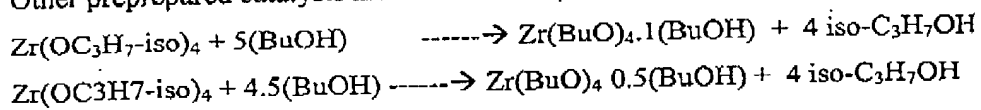
• **Applicant's Position:**

Applicant disagrees. On the contrary, the cited references neither disclose nor even suggest the presently claimed invention. The Applicant asserts that the Examiner has misread the cited references as well as the instant application.

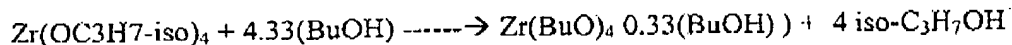
The invention provides a surprisingly advantageous process to produce linear oligomers from ethylene using a catalytic system that controls the size of linear oligomers to a range of four carbon atoms to 24 carbon atoms. This advantageous control is neither disclosed nor suggested by the combined references to Langer, Shiraki and/or Young. In fact, the cited disclosures are directed to mostly very short C4 to C8 oligomers and are silent as to producing any major size portion of linear C10 to C20 at high yield. Moreover, even one of ordinary skill in this art of catalytic conversion of ethylene would know from practical experience that the method of generating the catalytic systems alleged by the combined references would not result in the desirable size of alpha olefins demonstrated by the instant examples. It is also well understood that this art is so unpredictable that the combined cited method would not even remotely arrive at the claimed procedure. The presently claimed procedure in fact has been shown to generate a consistently favorable alpha olefin distribution of C4 to C20.

In addition to the previously submitted remarks in traverse of the Examiner's contentions and hindsight suppositions, Applicant asserts that the surprising advantages of the instant process is completely different from  $Zr(OBu)_4$  catalyst used by the primary reference. It has been unexpectedly found that the proper exchange does not even take place by mere mixing of alcohol and zirconium tetralkoxide as alleged. On the contrary, the catalyst used in the present examples is importantly prepared in the form of, e.g.  $Zr(OBu)_4(BuOH)$  being in an association at a ratio of 1:0.33 (Example 5) to 1:1.3 (Example 7), or preferably 1:1.23. The catalyst used in the present invention can be described as  $Zr(OBu)_4(BuOH)_x$  where  $x = 0.33-2.3$  and is added as a pre-prepared catalyst as is apparent from the protocols disclosed in the instant examples.. This is different from the  $Zr(OBu)_4$  catalyst used in the Langer reference.

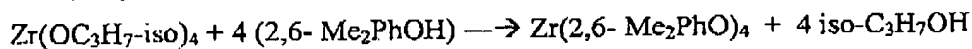
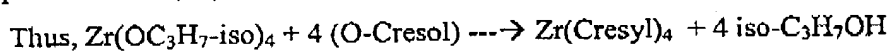
Other preprepared catalysts include for example



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All the above catalyst compositions containing a predetermined free alcohol content were employed in the oligomerization runs in the invention. The same method was employed for the preparation of Zr(IV) cresylate and Zr(IV) 2,6 dimethyl phenoxide.



The catalysts of the invention have a definite stoichiometry and cannot be generated even by a combination of 'in situ' methods described by Langer / Young / Shiraki. Considering the low level of predictability in the field of catalyst chemistry, it would be difficult to predict from the prior art whether dimers, oligomers or a polymer can be produced by merely employing transition metal alkoxide and an alkylaluminum. This is not obvious even to the person skilled in the art. One skilled artisan in this field would know that active catalytic species can be formed by a number different ways viz.

1. By reaction of M(OR)<sub>4</sub> complex with a Lewis acid (AlR<sub>3</sub>, RMGX)
2. Complexation of M(OR)<sub>4</sub> with Lewis acid (XnAlRn, BF<sub>3</sub> etc)
3. Use of above catalyst systems modified by electron donor or acceptor ligands such as, oxygenated / sulphur / phosphorous compounds.

Even with the same alkyl aluminium but varying the transition metal, different products are formed, as for e.g.,

Zr(OBu) <sub>4</sub> / AlEt <sub>3</sub>	1-Butene (Dimers)
Ti(OR) <sub>4</sub> / AlEt <sub>3</sub>	1-Butene + 1-C <sub>6</sub> + 1-C <sub>8</sub> + 1-C <sub>10</sub> (Oligomers) and NO POLYMER
Ti(OEt) <sub>4</sub> / EtAlCl <sub>2</sub>	70% Polymer and 30% Oligomer
TiCl <sub>4</sub> / EtAlCl <sub>2</sub>	92% oligomer (NO DIMER)

The following references support the above submissions:

1. Alkene & Alkyne oligomerization & related reactions, in Comprehensive Organomet. Chem., W. Keim, A. Behr, M. Roper Vol. 8, pp. 376-378.

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2. Adv. in Polymer Sci., Vol. 15, p.1 (1974). G. Olive & S. Olive.
3. H. Martin, Angew. Chem., 68, 306 (1956)
4. D.Jones, K.Cavell and W.Keim, J.Molecular Catalysis 138 (1999) 37-52)

support the argument that catalyst systems generated from preformed complexes are quite different in terms of activity and distribution of products in the oligomerization of ethylene. In this reference the authors compare  $ZrCl_2L_2$  (preformed catalyst) and  $ZrCl_4 \cdot 2HL$  (in situ system). The major observation is that preformed catalysts are more active than those formed in situ or bis-ligand adducts,  $ZrCl_4 \cdot 2HL$ . Activities of up to 60,000 turnovers/h were obtained with some complexes. In situ mixtures generally gave a much narrower oligomer distribution, with up to 95% of the oligomers occurring in the  $C_4$ - $C_{10}$  range for a number of the systems tested. Applicant offers to make these references available to the Examiner upon request. From the above it is clear that the performance of pre-prepared catalysts is quite different from those of in situ prepared catalysts. The teaching in the art is also in this direction. There is nothing in any of the citations relied on by the Examiner that identical results would be obtained whether the catalyst is used as pre-prepared catalyst or generated in situ. There is also no guidance or teaching towards the specific combination of a pre-prepared zirconium based catalyst with an alkyl aluminium and/or halide component.

Moreover, Applicant asserts that a mere broad knowledge on Ziegler-Natta Catalyst is insufficient to predict the reaction of a particular catalyst system under a given set of co-catalyst combination and operating conditions.

In addition to the arguments above, please reiterate the arguments used in the earlier response.

As would be understood by one of ordinary skill in the art, there is no assumption of predictability in catalyst systems such as those disclosed in the Langer reference and claimed in the instant invention. The Langer reference simply does not provide any teaching or suggestion that a halide ligand-containing zirconium complex which is generated *in situ* is equivalent to a non-halide containing zirconium complex which is not generated *in situ*. Nor would one of ordinary skill in the art *a priori* have any basis for equating the same.

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The very unpredictability of such catalyst systems is illustrated in Run 1 of Table IV of the Langer reference. As seen,  $\text{AlEt}_2\text{Cl} + \text{Zr}(\text{OPr})_4$  was inactive, while  $\text{AlEt}_2\text{Cl} + \text{AlEtCl}_2 + \text{Zr}(\text{OPr})_2$  in a mole ratio of 0.2:0.6:0.1 was evidenced to be active.  $\text{Zr}(\text{OBu})_4 \cdot \text{BuOH}$  exists as a trimer similar to  $\text{Zr}(\text{OPr})_4$  (Ref: Bradley *et al.*, *Metal alkoxide*, Academic Press, p. 63, 67, 1978). However the presence of toluene and similar solvents, the complex is in the form of a dimer (Ref: Vaartstra, *et al.*, *Alcohol adducts of alkoxides: Intramolecular hydrogen bonding as a general structural feature*, *Inorg. Chem.*, 1990, 29, 3126 - 3131). This dimer molecule is considered by the skilled artisan as different from  $\text{Zr}(\text{OBu})_4$ . It is also understood in the art that alcohol free  $[\text{Zr}(\text{OPr})_4]_m$  and  $\text{Zr}(\text{OPr})_4 \cdot \text{PrOH}$  have different properties. It therefore follows that in situ generated  $\text{ZrCl}_4 + n\text{-BuOH}$  is different from the defined  $\text{Zr}(\text{OBu})_4 \cdot \text{XbuOH}$  (where  $X = 0.3, 0.4, 1$ ) (a halide-free zirconium metal complex as used in the present invention) are different. In short, there simply is no predictability in the catalyst system.

The catalyst system of the Langer reference does not employ either  $\text{AlEt}_2\text{Cl}$  and/or  $\text{AlEtCl}_2$  as used in the Langer reference. Rather compounds such as  $\text{Et}_3\text{Al}$  and/or  $\text{Et}_3\text{Al}_2\text{Cl}$  are employed. Thus in their very constituents, the two catalyst systems are different.

Applicant submits that the present amendment of the claims places the application in condition for allowance. In particular, the order and ingredients of the instant procedure are more detailed. Newly added claims 31 and 32 are directed to the preferred embodiment of the instant process. No new matter has been introduced since the claimed subject matter is fully supported throughout the specification, especially on pages 3-5 as well as the examples and tables of the experimental results.

In this context Applicant asserts the patentable differences between the teachings of the Langer reference and the present invention which are set forth *supra*. Similarly, in regard to the Young reference the compounds taught therein also are known to have significantly different properties than those used in the present invention. For example, the Young reference teaches adducts of  $\text{ZrCl}_4$  with ketones, esters, nitriles, etc. This is a different class of compounds altogether from  $\text{Zr}(\text{OR})_4$  (e.g.,  $R = \text{butyl, phenyl}$ ). Applicant emphasizes that in fact the catalysts of the Young reference and those of the instant invention belong to different class of compounds



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and give a different reactivity. Moreover, the Young reference discloses aluminium compounds strictly for use with a catalyst having an adduct of zirconium tetrahalide as a catalytic component, in complete contrast to the claimed invention. Consequently, there is no motivation to combine the teachings of the Young reference with those of the Langer reference.

Furthermore, the Shiraki reference differs from the amended claims in that the cited catalyst system is based on the  $ZrCl_4$  system such that the catalyst system, as in Langer, is generated *in situ*. In the Shiraki discloses a method where  $ZrCl_4$  and alkylaluminiums are reacted first to provide a bridging complex. Upon adding butanol it bonds  $Zr-Cl$  and  $Al-C_2H_5$  simultaneously. Thus, reactivity of alkylaluminium with alcohol will form aluminum alkoxides (See, e.g., Kunicki et al, *Studies on the reaction of triethylaluminium with methyl alcohol*, Bull. Polish Acad. Sci. Chem., 33 No. 5 — 6, 1985). Thus, one of ordinary skill in the art would not be able to identify the identity of the active species for the oligomerization reaction. Finally, in complete contrast to the Examiner's contention, the Shiraki reference admits that thiophene is known at the time of the reference not to be a useful additive for enhancing the oligomer purity since sulfur compounds such as thiophene will cause contamination of the alpha oligomers. In point of fact, Shiraki teaches away from using such additives in order specifically teaches away from the use of thiophene in the system by replacing the sulfur compound by an alcohol (col 1 lines 29 — 35).

In light of such arguments with respect to the non-obviousness of the embodiments of the invention presently asserted, it is respectfully asserted that such 35 U.S.C. §103 rejections should be withdrawn with respect to claims 1 — 3, 4 — 12, 13 — 17, 18, 19, 20 — 22, 24 — 27, 29 and 30. Applicants respectfully assert that all pending claims in this application are in condition of allowance and are not taught, suggested, or made obvious by the prior art of record. Applicants respectfully request an early notice of allowance with respect to all claims that are presently pending.

There is no motivation seen in the cited references, taken alone or combined, to invent an ecologically friendly process such as the instant system presently claimed, which uniquely enables the use of inert and therefore unreactive and generally nontoxic solvents and catalysts. Moreover, the process affords energy saving temperatures and economic solvents less

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likely to endanger the environment with toxic waste. As such Applicant asserts that the claimed process would fill a long felt need for cleaner operating systems in the chemical industry.

Applicant asserts that the rejections of the claims, as presently amended, under 35 USC 103 are deemed improper and should be withdrawn, which favorable action is solicited.

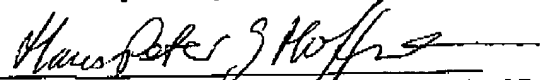
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### CONCLUSIONS

In view of the above, it is respectfully submitted that the subject matter of the pending claims which were rejected under 35 U.S.C. §§ 103(a), are patentable over the references of record. An early notice of allowance is therefore earnestly requested.

Respectfully submitted,

Date: April 2th, 2004



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